THE FORMATION OF SIDE BRANCHES OF DENDRITIC ICE CRYSTALS GROWING FROM VAPOR AND SOLUTION

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Dendritic ice crystals have been grown from the vapor and sucrose solution. The formation mechanism and the spacing of side branches have been studied. It was found that the formation mechanism of the side branches of dendritic ice crystals grown from the vapor was different from that from solution.

1. Introduction

The study of dendritic ice crystals is one of the important problems in the field of cloud physics, crystal growth and pattern formation. For example, almost all snow crystals that precipitate in nature are snowflakes composed of dendritic ice crystals [1]. On the other hand, although polyhedral crystals have been mainly studied in the field of crystal growth, recently, dendritic crystals with complicated form have attracted special interest [2–5] together with a study of form of fractal and DLA crystals [6,7]. The formation mechanism and the spacing of the side branches of dendritic ice crystals grown from the vapor and from sucrose solution are described in this paper.

2. Experimental apparatus

Fig. 1 shows the experimental apparatus for the growth and in situ observation of dendritic ice crystals. The apparatus is composed of a growth chamber (a) and its temperature regulators (b, c, d), a vacuum system (f, i) and an optical system (j, k, l, m, n). An ice sheet for water vapor supply and a growth substrate (sapphire) in the chamber were cooled down to about −15°C using thermoelectric modules. After the air in the chamber had been evacuated to about 10 Pa, water vapor was supplied by keeping the ice sheet at a slightly higher temperature than that of the growth substrate. After that, a small amount of diluted silver iodide smoke was inserted into the chamber.

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Fig. 1. Experimental apparatus for the formation and in situ observation of dendritic ice crystals. (a) growth chamber, (b) thermostatic refrigerated bath, (c) DC power supply, (d) temperature controller, (e) digital thermometer, (f) mercury manometer, (g) air inlet, (h) silver iodide smoke, (i) cold trap and vacuum pump, (j) video monitor, (k) video recorder, (l) video timer, (m) camera control unit, (n) video camera and microscope, (o, p, q) thermoelectric couples, (r) micro-computer.
together with clean air at $1.0 \times 10^5$ Pa. A minute ice crystal nucleated in air fell onto the growth substrate and was grown in air at $1.0 \times 10^5$ Pa at $-15^\circ$ C and a controlled supersaturation. Dendritic ice crystals were observed using a transmitted microscope (n) and recorded by a video camera. The growth chamber was also used for dendritic ice crystals grown from the sucrose solution. Before the operation of this chamber, a Teflon ring of 3.0 mm in inner diameter and 0.5 mm in thickness was placed on the growth substrate. The sucrose solution with freezing point of $-12.5^\circ$ C was poured into the teflon ring and a glass cover was mounted on it. A large ice crystal produced by keeping the sucrose solution at a slightly lower temperature than the freezing point was slowly dissolved. When a circular ice crystal of several 10 $\mu$m in diameter was formed, the ice crystal was grown again.

3. Experimental results

Fig. 2 shows a time sequence of a dendritic ice crystal grown in air at $1.0 \times 10^5$ Pa at $-15.5^\circ$ C. Photographs (a)–(e) are of an ice crystal grown at water saturation. In (a), the interfacial instability has already been generated on prismatic faces (arrow ↑). Six primary branches were formed as a result of interfacial instability of six prismatic faces (c). Although the tips of primary branches are initially off-faceted (c, d), facets were formed at the tips of them after time had elapsed (e). Photographs (f)–(h) are of an ice crystal at about 20% supersaturation over water. The side branches were formed at each corner of primary branches when the supersaturation increased (f), and the tips of primary branches became off-faceted again (f, g). Thereafter, facets were formed again at the tips of primary branches with time elapsed (h). Photograph (i) is of an ice crystal at about 40% supersaturation over water. Secondary side branches were formed again at each corner of primary branches when the supersaturation further increased (i). As shown in fig. 2, it is important for the formation of side branches of an ice crystal growing from the vapor that the facets are formed at the tips of primary branches and thereafter the supersaturation increases. Here, the spacing of side branches is given in fig. 2 (i).

Fig. 3 shows the formation process of a primary branch of a hexagonal plate grown in air at $1.0 \times 10^5$ Pa at $-15.0^\circ$ C and near water saturation. It is inferred that steps originated from two-dimensional nuclei nucleated at corners (1, 2) of a prismatic face successively advance to the center (3).
of the face. The central part (arrow ↑) of the prismatic face began to be unstable when advancing steps bunched with time elapsed (photo B). A primary branch was formed as a result of interfacial instability at two prismatic faces on both sides of the corner 1 (D, E, F). It is considered that the side branches in fig. 2 are formed by the same mechanism as that of the primary branches, the interfacial instability of two prismatic facets being formed at the tip of primary branches.

Fig. 4 shows a time sequence of a dendritic ice crystal grown in sucrose solution at −15°C and 2.5°C supercooling. The tips of primary branches of the ice crystal grown from the solution are constantly rounded. With solution growth, side branches are formed in a state of round tips, and the tips of primary and side branches are constantly rounded during the growth.

Fig. 5 shows the relationships between the spacing of side branches and the tip velocity of den-
Dendritic ice crystals grown from (a) vapor at different supersaturations and (b) solution at different supercoolings. Here, with ice crystals grown from solution, the spacing of side branches which can be measured were completely plotted. The spacing of side branches for solution growth is narrower than that for vapor growth. The spacing of side branches for solution growth decreases with increasing tip velocity. That is, the side branches sprout easily with increasing tip velocity when the supercooling increases. On the other hand, the spacing of side branches of ice crystals grown from the vapor increases with increasing tip velocity. That is, the side branches sprout with more difficulty with increasing tip velocity when the supersaturation increases. This means that the formation mechanism of side branches of dendritic ice crystals growing from the vapor is different from that which is operative in growth from solution.

4. Discussion and concluding remarks

Dendritic ice crystals were grown from the vapor and solution. The tips of the branches of ice crystals growing from the solution have constantly rounded regardless of the degree of supercooling. As seen in fig. 4 (e), infinitesimal side branches are formed at the tips of primary branches. It is inferred that the side branches of ice crystals growing from the solution are formed by the interfacial instability at the entire ends of primary branches. The spacing of side branches of ice crystals growing from the solution decreases with increasing tip velocity when the supercooling increases. On the other hand, the tips of primary branches of ice crystals growing from the vapor change from off-faceted to faceted and vice versa with decreasing or increasing supersaturation. In this case, the side branches are formed by the interfacial instability of prismatic facets formed at the tips of primary branches, that is, it is important for the formation of side branches that the tips of primary branches are transformed from off-faceted to faceted.

In conclusion, dendritic ice crystals growing from solution are dominated only by the volume diffusion process of water molecules onto ice crystal surfaces, while those from the vapor are dominated not only the volume diffusion process of water molecules onto ice crystal surface, but also by the kinetic process of water molecules on ice crystal surface.

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References